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NIF TARGET CAPSULE WALL AND HOHLRAUM TRANSFER GAS EFFECTS ON DEUTERIUM-TRITIUM REDISTRIBUTION RATES

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Abstract

The effects of temperature and age on the times required for beta-heating-induced redistribution of a 50-50 mole percent mixture of deuterium and tritium (DT) in a spherical capsule are investigated analytically and numerically. The derivation of an analytical solution for the redistribution time in a one-dimensional binary diffusion model, which includes the capsule thermal resistance, is first described. This result shows that the redistribution time for a high conductivity capsule wall is approximately doubled after 8 days of ^3He formation. In contrast, with a low thermal conductivity capsule wall (e.g., polyimide), the redistribution time would increase by less than 10%.

The substantial effect of the capsule wall resistance suggested that the resistance to heat transfer from the capsule through the surrounding transfer gas to the hohlraum wall would also influence the redistribution process. This was investigated with a spherical model, which was based on accounting for energy transfer by diffusion with a conduction heat transfer approximation. This made it possible to solve for the continuous temperature distribution throughout the capsule and surrounding gas. As with the capsule the redistribution times depended on the relative values of the thermal resistances of the vapor, the capsule, and the transfer gas. With increasing thermal resistance of the vapor (increased concentration of ^3He) redistributions times for hydrocarbon capsules were significantly less than predicted by the one-dimensional model, which included the capsule wall resistance. In particular for low ^3He concentrations the time constant was approximately 10% less than the minimum one-dimensional value of 27 minutes. Further analytical and experimental investigation focused on defining the relations between the thermal resistances under which the one-dimensional model analysis applies is recommended.

Introduction

The targets for direct or indirect inertial confinement fusion will be spherical shells of solid fuel on the inside of thin walled spheres. The fuel will be a 50-50 mole percent mixture of deuterium and tritium (DT). The thickness of the shells, of both the fuel and surrounding

capsules, will have to be very uniform. Current plans for producing such targets are to fill a thin-walled plastic or metal spherical shell with DT gas through a small fill tube, and then cooling the shell and gas until the DT solidifies around the inner surface. During cooling liquid will condense around the inner spherical surface and flow to the bottom. Continued cooling will result in a solid layer, which is much thicker on the bottom than on the top. Tritium, however, is radioactive, and as it decays to a ^3He atom, it emits a beta particle and an antineutrino. The ^3He atoms tend to escape into the vapor region. Since the betas are absorbed within a distance of approximately 10 microns, liquid or solid tritium has effectively a nearly uniform internal heat generation rate. This leads to a temperature distribution, which increases quadratically with distance from a cooled boundary. Since the temperature rise is greater in a thicker than in a thinner layer, the temperature of the inner surface of the thicker layer on the bottom will be higher than that at the inner surface at the top. This will cause sublimation at the lower surface and diffusion of the vapor to and condensation on the upper surface. This redistribution mechanism (which has been termed beta layering) should produce a solid spherical layer of uniform thickness inside the containing shell.

The above spontaneous redistribution process suggested by Miller¹ has been demonstrated by Hoffer and Foreman² in cylinders. Redistribution was observed to decrease at an exponential rate with time. The time constant τ for this rate was in good agreement with the value predicted by Martin et al³ from a one-dimensional model (see Figs. 1 and 2) for DT that is relatively free of the beta-decay ^3He atoms. They also showed that τ increased with the age of the DT, because of the buildup of ^3He through which the DT must diffuse, and that τ increases strongly with decreasing temperature. A quantitative result for the time constant of aging DT was developed by Bernat et al⁴ based on assuming the thermal conductivity of the capsule was infinite. Predictions for τ were about 30 to 50 percent lower than experimental measurements reported for DT in a relatively large sapphire sphere (15 mm in outside diameter).

The objectives of the present study were 1) to extend the one-dimensional analysis referenced above to include the effect of the thermal resistance of the capsule wall, and 2) to develop a spherical model which accounts for the energy transfer associated with the DT diffusion, and permits numerical calculation of the temperature field throughout a hohlraum.

One-dimensional Analysis

When the DT vapor in a target sphere is cooled and condenses, most of the liquid falls to the bottom with the rest clinging to the upper inner surface of the capsule. With further cooling this distribution becomes solid. As a first approximation the solid region can be modeled as the volume surrounding an eccentric sphere as illustrated in Fig. 1. The rate of sublimation and

transfer of DT will be proportional to the greater thickness around the bottom half. We then assume that what occurs at this location will be representative of the process and that the redistribution of DT in a small region around the vertical axis is one-dimensional. The temperature distribution along this axis will vary as shown on the one-dimensional model of Fig. 2. The maximum temperature occurs just inside the inner surface of the thicker layer. Heat generation to the right of (above) this location causes sublimation of DT, which then diffuses through the ^3He and condenses at the top. It will be shown later that this is the primary mechanism for energy transfer until the concentration of ^3He approaches and exceeds that of DT.

All of the ^3He produced in the solid region is assumed to escape into the vapor. Since the concentration of ^3He builds up slowly, conditions in the vapor (Fig. 2) can be considered quasi-steady at any time. Although the DT gas is made up of molecules of D_2 , DT, and T_2 , they are similar and can be modeled as a single species. Then regarding ^3He as a second species, the rate of transfer of DT at any time from the thicker to the thinner layer can be approximated as binary diffusion of DT through ^3He , where the net flux of ^3He is zero. These are the conditions assumed in the following analysis.

Denoting the molecular concentration of DT by n_{DT} and that of ^3He by n_3 , the applicable diffusion equations are

$$-D_{DT3} \frac{dn_{DT}}{dx} + n_{DT}v = \dot{n}_{DT} \quad (1)$$

$$-D_{DT3} \frac{dn_3}{dx} + n_3v = 0 \quad (2)$$

in which D_{DT3} is the binary diffusion coefficient for DT diffusing through ^3He , and v is the local mass average velocity. Generation of ^3He in the low-density vapor is neglected. Eliminating v yields

$$\dot{n}_{DT} = D_{DT3} \left[\frac{n_{DT}}{n_3} \frac{dn_3}{dx} - \frac{dn_{DT}}{dx} \right] \quad (3)$$

The temperature difference which develops between the inner surfaces of the thicker and thinner DT layers in capsules ~ 2.0 mm in diameter is only on the order of a millikelvin, and the pressure through the vapor region will be essentially uniform. At the pressures and temperatures involved the vapor obeys the perfect gas equation of state⁵. The molecular concentration $n = n_{DT} + n_3$ can

be considered constant^a and $dn_3/dx = -dn_{DT}/dx$. Making this change for dn_3/dx in Eq. (3) and rearranging yields the following relation for $\dot{n}_{DT}dx$:

$$\dot{n}_{DT}dx = Dn \left[\frac{d(n - n_{DT})}{(n - n_{DT})} \right] \quad (4)$$

We now want to express the DT diffusion rate \dot{n}_{DT} as a function of the properties at the inner surfaces of the thick and thin layers. This is achieved by integrating Eq. (4) and requiring the result to match the heat rates and temperatures at the inner surfaces of the solid DT layers (x_1 and x_2 in Fig. 2). In carrying out this integration it will be assumed that the variations of the diffusion coefficient D_{DT3} and the molecular concentration n are negligibly small so that mean average values can be used. For the non-polar pair DT and ^3He , D_{DT3} is almost independent of composition (see Appendix). Although it does depend on pressure and temperature, the pressure in this application is constant and the temperature variation is small (on the order of mille Kelvin for small spheres and tens of mille Kelvin for spheres of 10 to 15 mm in diameter). The small temperature variation also justifies considering n constant in this integration. The result evaluated at x_1 and x_2 is then

$$\dot{n}_{DT} = \frac{D_{DT3}n}{S} \ln \left[\frac{n - n_{DTV}(x_2)}{n - n_{DTV}(x_1)} \right] \quad (5)$$

where $S = x_2 - x_1$. The DT vapor at x_1 and x_2 (now denoted by n_{DTV}) is assumed to be in the saturated state at the respective surface temperature.

The surface temperatures at x_1 and x_2 are determined by the resistance to the flow of the heat released during tritium decay to the hohlraum cooling rings. In order to investigate the interaction between the vapor, the solid DT, the capsule and the transfer gas it is necessary to have the diffusion rate expressed in terms of the temperatures at the inner surfaces of the thick and thin layers. Since the temperature difference between these two saturated states is very small, it is possible to approximate the result for \dot{n}_{DT} as a function of this temperature difference by expanding the logarithm term in Eq. (5) in a Taylor series about x_1 . Dropping all terms of this series but the first, replacing dn_{DTV}/dx by $(dn_{DTV}/dT)(dT/dx)$, and approximating dT/dx by $(T_2 - T_1)/(x_2 - x_1)$ gives^b

^a To illustrate the error involved assume $T_1 = 19.5$ K, and a decrease to 19.495 at T_2 . At x_1 $n = 7.53066 \times 10^{25} \text{ m}^{-3}$, which is the sum of n_{DTV} and n_3 . At x_2 $n_{DTV} = 6.43591 \times 10^{25} \text{ m}^{-3}$ and $n_3 = 1.09564 \times 10^{25} \text{ m}^{-3}$ if n is constant and $1.09761 \times 10^{25} \text{ m}^{-3}$ if p is constant. The difference is about 0.2% of n .

^b The approximation for \dot{n}_{DT} in Eq.(6) becomes more accurate with time because as the concentration of ^3He increases, the argument of the logarithm term decreases and the one-term series approximation becomes more

$$\dot{n}_{DT} = \frac{Dn}{S} \ln \left[\frac{n - n_{DTV}(x_2)}{n - n_{DTV}(x_1)} \right] = \frac{Dn}{S} \left[\frac{n_{DTV}(x_1) + dn_{DTV}(T_1)/dT}{n_3(x_1)} \right] (T_1 - T_2) \quad (6)$$

Introducing the empirical relation for the pressure of saturated DT vapor recommended in Ref. 5 in the ideal gas law, and differentiating this relation yields the derivative required in Eq. (6).

The temperature distribution in each DT layer is determined by requiring that the heat flux out of the thicker layer and into the thinner layer be equal to the product of the DT diffusion rate \dot{n}_{DT} and the heat of sublimation h_s . This heat flux is one of the two boundary conditions, which must be satisfied by the temperature distribution in each layer (ℓ_1 and ℓ_2) and the capsule wall thickness ℓ_c . The other is the outer surface of the capsule wall, which will be specified to be at a uniform temperature T_0 . For these boundary conditions, constant thermal conductivities, k_s and k_c of the solid DT and the capsule wall respectively, and heat generation rate G per unit volume of the solid DT, the temperature difference between the inner surfaces is found to be

$$T_1 - T_2 = L \left[\frac{G(\ell_1 - \ell_2) - 2\dot{n}_{DT}h_s}{2k_s} \right] + \ell_c \left[\frac{G(\ell_1 - \ell_2) - 2\dot{n}_{DT}h_s}{k_c} \right] \quad (7)$$

Substituting this result for $(T_1 - T_2)$ in Eq. (6) and rearranging yields the following relation for \dot{n}_{DT} :

$$\dot{n}_{DT} = \frac{(\ell_1 - L/2)}{\left[\frac{n_3(x_1)k_s k_c S}{D_{DT3}nG(dn_{DT}(T_1)/dT)(Lk_c + 2\ell_c k_s)} \right] + h_s/G} \quad (8)$$

A redistribution time constant is determined from this explicit relationship by first recognizing that the negative of the rate of change of the thicker layer, $-d\ell_1/dt$, multiplied by its molecular density is equal to \dot{n}_{DT} ; i.e.,

$$-\rho_s \frac{d\ell_1}{dt} = \dot{n}_{DT} = \frac{(\ell_1 - \frac{L}{2})}{N} \quad (9)$$

when $L - \ell_1$ is substituted for ℓ_2 and N denotes the denominator in Eq.(8). Next integrating from an arbitrary initial value ℓ_{10} to ℓ_1 gives

accurate. At 18 K, for example, after 1 day of ^3He formation in the 15 mm capsule to be discussed later, \dot{n}_{DT} calculated from Eq.(6) is about 12% higher than the exact value determined from Eq. (6); after 10 days \dot{n}_{DT} is about 5.5% higher, and by 20 days the difference has decreased to about 4%. However, substituting for $(T_1 - T_2)$ from Eq. (7) in Eq. (6) compensates for this error since \dot{n}_{DT} occurs in Eq.(7). As a result \dot{n}_{DT} determined from Eq. (10) is accurate to about $\pm 4\%$, which is acceptable for this analysis.

$$\frac{\ell_1 - \frac{L}{2}}{\ell_{10} - \frac{L}{2}} = \exp \left[\frac{-t}{\rho_s N} \right] \quad (10)$$

This result indicates that the DT redistribution process varies exponentially with a time constant τ equal to $\rho_s N$. The complete expression,

$$\tau = \frac{\rho_s S n_3 k_c k_s}{D_{DT3} G n (dn_{DTV}(T)/dT) (L k_c + 2 \ell_c k_s)} + \frac{\rho_s h_s}{G} \quad (11)$$

shows that τ is directly proportional to n_3 , which increases linearly with time (for times less than about one year) due to beta decay. If no ^3He has formed, $\tau = \rho_s h_s / G$, which is equal to 26.9 minutes for the 50-50 DT mixture. The other parameters on which τ depends (except S and L) vary with temperature. The variation is dominated by the derivative of the saturated DT vapor density, which depends more strongly on temperature than the other properties.

The variation predicted by Eq. (11) for a temperature of 18.5 K is plotted in Fig. 3 as a function of the capsule thermal conductivity^c. The dramatic effect of the wall material is apparent, particularly as the time for ^3He formation increases. Changing the capsule from copper-doped beryllium to polyimide reduces τ by as much as one half after 8 days of ^3He production. This is a very significant effect since the total time for redistribution will be four to five times τ . Also note that for the particular case considered in Ref. 4, where the capsule conductivity $k_c \rightarrow \infty$, Eq. (11) reduces to

$$\tau_{k_c=\infty} = \frac{\rho_s k_s S n_3}{D_{DT3} n G L (dn_{DTV}(T)/dT)} + \frac{\rho_s h_s}{G} \quad (12)$$

This relation gives the asymptotic values approached by the curves in Fig. 3.

Comparison with 15 mm Sphere Measurements -- Limited results for the effect of age on the redistribution time for DT in a 15 mm OD sphere were reported in Ref 4. Absolute magnitudes for τ varied linearly with increasing n_3 but appeared to converge on a value of about 40 minutes instead of the predicted 26.9 minutes calculated from $\rho_s h_s / G$. The rates of increase with aging time for the three test temperatures of 17.2, 18.0, and 18.9 K are plotted in Fig. 4. An analytical relation was also developed in Ref 4 for predicting the time constant increase rate. This is represented by the upper curve, which can be seen to be about 50% higher than the data. As is pointed out in the Appendix this difference is probably due to the property values used in

^c Property values for DT were taken from Ref. 5, except for k_s , which were obtained from the more recent study of Collins et al⁶. Prediction of the diffusion coefficient D_{DT3} is discussed in the Appendix.

calculating the diffusion coefficient. The middle curve in Fig. 4, which agrees quite well with the data, was determined using the Hirschfelder et al¹⁰ equation (see Appendix) for the diffusion coefficient. The deviation of the data point for 17.2 K can be attributed to the uncertainty in the measurements. Values of τ predicted by either Eq. (11) or Eq. (12) were in similar agreement with the experimental data. The rapid increase of τ'_{DT} with decreasing temperature is primarily attributable to the decrease in the rate of change of the density of saturated DT vapor with temperature.

Comparison with Simpson, Hoffer and Foreman Measurements¹⁴—A series of time constant measurements reported by Simpson et al were made with an 8.776 mm OD and 6.582 mm ID polycarbonate sphere. The wall was over 1.0 mm thick and the material a very poor conductor (a thermal conductivity on the order of 0.00005 W/mm K – Ref. 15). These are the conditions for which Eq. (11) is expected to apply. The lower curve in Fig. 4 represents the derivative with respect to time of the time constant τ defined by Eq. (11). The Simpson et al tests were conducted with charges of DT equivalent to uniform layers of 200 μ m and 650 μ m. Time constant data for the 650- μ m thick layer exhibited the characteristic behavior of τ increasing approximately linearly with time for ³He formation, see Figure 8. The slope of their curve was measured to be about 0.56 minute per day. This value is only about one half the 1.0-minute per day increase predicted by the gradient of Eq. (11) for the rate of change of τ . The lower experimental value is postulated to be due to the resistance of the very low conductivity capsule wall and the transfer gas surrounding the capsule -- similar to the low value for τ calculated for the “Ch – lo-k vapor” temperature distribution shown in Fig.7.

Influence of Spherical Geometry and Transfer Gas on DT Redistribution -- The heat flow from the DT in the one-dimensional model is assumed to flow radially into the surrounding transfer gas. The flow however, is more complex. It has been shown to depend on the relative magnitudes of the thermal resistances of the DT layers and the capsule wall. Variation with the angle from the bottom of the capsule can be anticipated due to the change in thickness of the DT layer. A circumferential temperature gradient will develop in the capsule wall, and this could influence the one-dimensional flow assumed in the above analysis. The increasing area for heat flow with increasing radius in a sphere may also have a small effect. These factors suggested that the overall heat flow from a capsule through the transfer gas to the hohlraum cooling rings should be considered. This would include the possible influence of heat released by tritium in the vapor. Although the tritium density in the vapor is very low compared to that in the solid, the volume of the vapor is substantially larger, and the distance over which heating occurs is relatively long.

Numerical solution Model

A simplified model suitable for numerical solution was developed for investigating the overall pattern of heat transfer in and from a capsule mounted in a hohlraum. The key new feature was to express energy transfer by diffusion through the vapor in the capsule in terms of an effective thermal conductivity. In this way a continuous solution for the temperature and heat flux distributions in the capsule wall and the solid and vapor DT regions could be obtained with a finite element conduction program.

Energy Transfer by Diffusion -- Fundamentally the redistribution of DT is a transient process with coupled conduction heat transfer and diffusion. The process is relatively slow, however, so that quasi-steady-state conditions can be assumed. The local diffusion rate \dot{n}_{DT} of DT vapor at any x is given by Eq.(3). When dn_3 / dx is replaced by $-dn_{DT} / dx$, $n_{DT} + n_3$ by n , and dn_{DT} / dx by $(dn_{DT} / dT)(dT / dx)$ we obtain

$$\dot{n}_{DT} = -\frac{D_{DT3}n}{n_3(x)} \left\{ \frac{dn_{DTV}(T)}{dT} \right\} \frac{dT}{dx} \quad (13)$$

in which the subscript DTV indicates the assumption^d that the DT vapor is saturated or very close to the saturated vapor state as it diffuses from x_1 to x_2 and changes from T_1 to T_2 . Multiplying the DT diffusion rate \dot{n}_{DT} times the heat of sublimation h_s yields the local energy transfer rate by diffusion q_d as

$$q_d = \dot{n}_{DT}h_s = -\left[\frac{D_{DT3}nh_s}{n_3(x)} \left\{ \frac{dn_{DTV}(T)}{dT} \right\} \right] \frac{dT}{dx} \quad (14)$$

Observe that this equation can be expressed as

$$q_d = -k_d \left(\frac{dT}{dx} \right) \quad (15)$$

in which k_d represents

$$k_d = \left[\frac{D_{DT3}nh_s}{n_3(x)} \left\{ \frac{dn_{DTV}(T)}{dT} \right\} \right] \quad (16)$$

This indicates that k_d can be defined as an effective thermal conductivity for diffusion. Also note that the molecular fluxes and gradients and velocities in Eqs.(1) and (2) are vector quantities. Hence if more than one dimension is involved, a similar set of equations would apply for each dimension, and from which equations similar to Eq.(14) for the fluxes could be developed.

Considering convective transport in the vapor to be negligible, energy transfer will be by

^d This assumption is justified by noting that 1) the vapor is saturated at x_1 and x_2 , 2) temperature and pressure changes are small, and 3) there is no heat removal and heat addition is very small.

molecular thermal conduction and diffusion. Assuming these two mechanisms to be independent, the energy transfer rate will be determined by a total vapor conductivity k_v equal to the sum of the thermal conductivity k_c and the diffusive conductivity k_d ; i.e., $k_v = k_c + k_d$. The advantage of introducing this approximation is that the continuous temperature distribution throughout the capsule wall, the DT solid and vapor regions, and the transfer gas can then be determined by solving the differential equation governing steady-state conduction with appropriate boundary conditions. The system can be considered axially symmetric.

Thermal and Diffusive Conductivities -- As defined by Eq. (16) k_d will vary locally. The change in the quantity $D_{DT3}nh_s$ will be negligible, and the variation of $dn_{DTV}(T)/dT$ over a representative maximum capsule temperature range of 0.010 K (in ~ 2 mm) is less than 1.0%. The percentage variation of $n_3(x)$ will be greater than this (5-10%) during the first few days of formation, but will decrease rapidly as the concentration approaches that of the DT vapor. Since these effects are minor, constant average values were used in evaluating k_d . Curves showing the variation of k_d and k_c for 19.5 K are plotted in Fig 5.

Values of k_d calculated from Eq. (16) after 1 day of ^3He formation were 0.0082 W/mm K at 17.2 K and 0.0238 W/mm K at 19.5 K. Since k_d varies inversely with n_3 , which increases linearly with time, after 5 days of aging at 17.2 K, k_d will decrease to 0.00164, and after 20 days to 0.000411 W/mm K. At 19.5 K the 5 day value will be 0.00477 and 0.00192 W/mm K at 20 days. The total thermal conductivity k_v of the DT vapor will be the sum of diffusion and conduction components. The conduction component will depend on the mole fractions of DT and ^3He , and the average cell temperature. The conductivity of ^3He was evaluated with the empirical relation given in Ref. 5^c. In view of its small magnitude an average value of 0.0000289 W/mm K (from 17.2 to 19.5 K in Table 3.1 of Ref. 5) was taken for DT. Weighted values for the DT vapor- ^3He mixture conductivity were then calculated based on molecular concentrations. For illustrative values of the vapor mixture thermal conductivity k_c consider a 2.22 mm OD capsule with a 0.080 mm solid DT layer. The ratio of V_v / V_s is on the order of 3.3 and the rate of increase of ^3He would be around 1.4×10^{24} atoms/m³ per day. At this rate the partial pressure of ^3He will equal that of DT at 17.2 K in about 10 days, and in about 30 days at 19.5 K. For this ^3He buildup rate values of the vapor conduction conductivity at 17.2 K will increase from around 0.00001 W/mm K at 5 days to 0.000017 in 20 days, and at 19.5 K from about 0.000012 to 0.0000164 W/mm K.

Comparing the above values shows that for aging times up to 5 days k_d will be from over 40 to over 250 times larger than the vapor thermal conductivity k_c . After 20 days it will still be from 20 to 10 times larger. Hence energy transfer up to 20 days will be dominated by diffusion.

^c Page 198: $k \approx 3.18 \times T^{0.7738}$ in W/mm K.

Natural convection -- For fusion targets up to 2 mm in diameter values of the Rayleigh number for the vapor region are found to be negligibly small compared to the value of 1700, above which natural convection may develop. However, for the measurements presented earlier for a 15 mm sphere with a 1.2 mm thick wall, the maximum temperature differences between the inner surfaces of the top and bottom DT layers calculated from the one-dimensional model were on the order of 0.06 K. At average cell temperatures ranging from 17 to 18.9 K and a vapor sphere diameter of 1.1 cm, the maximum Rayleigh number was estimated to vary from about 1400 to 6000 based on the thermal conductivity of the vapor. This indicates that some natural convection would occur at temperatures above about 17.5 K.. Since natural convection is not expected to occur in smaller cells, energy transfer in the vapor region was assumed to be by conduction and diffusion as described above.

Numerical Results

The one-dimensional system shown in Fig. 2 was first zoned with approximately 25 rectangular elements in the capsule wall, 100 in the solid DT layers, and 100 in the vapor region. Wall, solid DT, and vapor region thickness were selected to represent cells from 8 to 15 mm in diameter. Wall thickness of 0.5, 1.0 and 1.2 mm, and a thickness of the solid DT layers of 0.5 to 0.75 mm were assumed. Calculations were made with the thicker layer 3 and 9 times the thickness of the thinner layer. The heat generation rate in the solid was taken as 0.977 W/mole, which is equivalent to 0.0491 W/cm³ (Ref. 5). Quasi-steady-state temperature distributions were determined with the COSMOS⁶ finite element computer code. The output of this program includes values of the heat flux in each coordinate direction at each node, as well as the temperatures. A system time constant can be evaluated from the value of the heat flux at the inner surface of the thicker layer. This can be seen by assuming that the rate of change of thickness of this layer is given by

$$\ell_1 - L/2 = (\ell_{10} - L/2)\exp(-t/\tau) \quad (17)$$

Then

$$\frac{d\ell_1}{dt} = -(\ell_{10} - L/2)\exp(-t/\tau)/\tau \quad (18)$$

and, assuming $t=0$ for any arbitrary value of ℓ_{10} ,

$$\tau = \frac{(\ell_{10} - L/2)}{d\ell_1/dt} \quad (19)$$

Multiplying numerator and denominator of Eq.(19) by $\rho_s h_s$ then gives

$$\tau = \frac{(\ell_{10} - L/2)\rho_s h}{q_{\ell_1}} \quad (20)$$

where $q_{\ell_1} = -(d\ell_1/dt)\rho_s h_s$ denotes the heat flux at the inner surface of the thicker layer.

Results were first computed for an average DT layer thickness of 0.75 mm and vapor length of 11.1 mm. These dimensions were based on the size and DT loading of cells, which can be conveniently studied experimentally. A very high thermal conductivity capsule (such as sapphire) was assumed so that constant and equal wall temperatures could be specified as boundary conditions. When heat release in the vapor is not included, computed results for τ increased linearly with time. Hence τ' was constant for any average DT solid and vapor temperature level. Dividing the results for τ' by the appropriate value of $f_g = (V_s / V_v)(S / L)$ yielded values for τ'_{DT} which were in excellent agreement with predictions from the analytical solution, Eq.(12), thus validating the technique of representing diffusion as an energy transfer mechanism.

Effect of Heat Generation in Vapor Region -- To investigate the effect of tritium decay in the vapor region, the heat release (based on tritium concentration) was assumed to be spatially uniform and the temperature distribution for a one-dimensional model considered (see Appendix). One half of the heat released in the vapor is shown to be transferred to the x_1 interface (Fig. 2). Comparing this to the heat release rate in the subliming DT indicated that the effect on the heat flux at this location to be on the order of 3%. However, calculations of time constants from overall solutions for the hohlraum temperature distributions did not show this difference.

Hohlraum Model Results -- The behavior of an complete hohlraum system was then investigated by solving for the temperature distribution throughout a finite element model of the capsule, surrounding gas and hohlraum wall. A 2.22 mm OD capsule was mounted between two thin Formvar (polyvinyl) films at the center of a 9.75 mm high hohlraum. Natural convection in the transfer gas was essentially eliminated by installing two additional horizontal thin films in both the upper and lower halves of the hohlraum⁹. The final zoned configuration had seven separate gas regions. Temperature and heat flux distributions were obtained assuming conduction only with the COSMOS/M⁷ code. Representative results for the region enclosing the capsule are shown in Fig. 6 for a diffusive conductivity of 0.0035 W/mm K, determined from Eq.(16) for a mean temperature of 18.5 K and 5 days of ³He generation. The capsule wall and surrounding gas thermal conductivities were 0.00015 W/mm K (a hydrocarbon) and 0.000294 W/mm K respectively. The isotherms in the thick (bottom) DT layer indicate heat flow into the vapor region in the lower half of the capsule. This would tend to augment the diffusive energy flux along the vertical axis. Evaluation of a time constant from Eq. (20) using the average slope

of the lower one tenth of the nodal temperatures along the vertical axis (in the vapor) to calculate the heat flux from the lower surface yielded a value of 24 minutes. This is about 9% lower than the lower asymptotic value of 26.9 minutes in Fig. 4. This is due in part to the effect of the increasing volume of a DT layer with radius in a spherical shell. An estimate indicated that a decrease of about 7% from the one-dimensional value of τ (to about 25 minutes) is possible. The remaining difference could be due to radial inflow from the DT layer around the bottom in a capsule with a low conductivity wall. One might conclude that the difference between these two τ values is an indication of how accurately the one-dimensional model represents the redistribution process.

Additional solutions for the overall temperature distribution in a capsule-hohlraum system and calculation of time constants do not bear this out. Fig. 7 shows the temperature distributions along the vertical axes in hydrocarbon and beryllium capsules. With each material low and high values for the vapor thermal conductivity were specified (0.0013 and 0.013 W/mm K). The two curves for the hydrocarbon capsule show that, when the resistances of the capsule wall and the vapor are relatively high, the resistance of the transfer gas has a dramatic effect. Calculation of a time constant with Eq. (20) for 11 days of ^3He formation yields a value of 27.4 minutes. In contrast the one-dimensional model Eq. (11) gives 40 minutes. This trend is indicated in the series of time constant measurements reported in Ref. 14. These are reproduced in Fig. 8 and compared with the distributions from Eq. (11) and from a solution for the temperature distribution in a hohlraum simulating the system of Ref. 14. This comparison illustrates the limitation of the one-dimensional model and further substantiates the applicability of the diffusive thermal conductivity.

Conclusion

An equation based on a one-dimensional model of quasi-steady heat flow in the DT vapor, the solid layers and the capsule wall for predicting the time constant for the redistribution of DT in a millimeter sized capsule is derived. This relation clearly establishes the conclusion that the redistribution time depends on the relative magnitudes of the resistance to diffusion and heat transfer through the solid DT and the capsule wall.

The similarity between the relation for DT diffusion and conduction made it possible to define a thermal conductivity for diffusion k_d . By defining the conductivity of the vapor to include both diffusion and conduction the temperature distribution throughout the capsule, transfer gas and hohlraum was determined numerically. Time constant variation with time for ^3He formation determined from the hohlraum temperature distribution was in reasonable

agreement with limited experimental data. However, predictions based on the one-dimensional model of the vapor, DT layer and capsule wall over estimated values for τ , indicating it's limited applicability.

APPENDIX

Coefficient for Diffusion of DT Through ^3He

The observed redistribution of solid DT in a capsule produced by the decay of tritium atoms has been modeled as binary diffusion of DT molecules through a mixture of DT and ^3He gas. The key quantity involved in predicting the rate of transfer is the diffusion coefficient, which is denoted by D_{DT3} . Formulas for calculating the value of this coefficient are based on kinetic theory and the intermolecular force field between the two molecules involved. The basic relation for gases at low pressure is referred to as the Chapman-Enskog equation⁹. A particular form in which the molar density of the gas mixture is approximated by the ideal gas law, has been proposed by Hirschfelder, Bird and Spotz.¹⁰ This relation has been shown to yield results for the diffusion coefficient accurate to about 6%¹⁰. It will be denoted by Dh_{DT3} and is given as

$$Dh_{DT3} = 0.00183 \frac{\sqrt{T^3 \left(\frac{1}{M_{DT}} + \frac{1}{M_3} \right)}}{p \sigma_{DT3}^2 \Omega} \quad (1A)$$

The absolute temperature T is in Kelvin, M_{DT} , and M_3 are the molecular weights of DT and ^3He , p is the pressure in atmospheres, σ_{DT} in Angstroms, and $\Omega_{D,DT}$ a dimensionless function of temperature, are parameters in the Lennard-Jones intermolecular potential field for a molecule of DT and an atom of ^3He . These units give Dh_{DT3} in cm^2/s .

The predicted variation of Dh_{DT3} for the vapor mixture after 5 days and 20 days of ^3He formation in a 2.2 mm diameter capsule are plotted in Fig, 1a. Values for σ_{DT} and $\Omega_{D,DT}$ were obtained from Refs, 11 and 5. A second semi empirical relation proposed by Slattery and Bird is also plotted in Fig. 1a¹³. Their approach involves the application of the concept of corresponding states. Although results appear to be identical to the Hirschfelder et al curve, they are slightly less accurate.

For comparison the diffusivity was also calculated (as suggested In Ref 5, p. 35) bu

multiplying the self-diffusion coefficient of H_2 by a molecular weighting factor. $(M_{^3He} M_{DT}) / (M_{^3He} + M_{DT})$. The k_s values for solid DT were taken from the more recent study of Collins et al.⁶ The results (see lower curve in Fig. A1) are about 40-50% less than the upper curve. Although it would appear that the semi empirical relation for diffusivity is more accurate, additional experimental data are needed to support this conclusion.

Effect of Heat Release in Vapor

Assuming that heat is released uniformly in the DT vapor region of a capsule, the volumetric heating would be given by

$$G_{DTV} = n_{DT} R_d E \quad (2A)$$

in which R_d denotes the tritium decay rate in atoms per second per atom and E is the energy released per decay in Joules. With a vapor temperature of 19.5, for example, $G_{DTV} = 105 \text{ W/m}^3$.

The magnitude of the effect that this can have on the DT redistribution time can be estimated by determining the change introduced in the one-dimensional model of Fig.1. The general solution for the temperature distribution for conduction heat transfer through the vapor from x_1 to x_2 with uniform volumetric heat generation is

$$T - T_1 = \frac{G_{DTV} x^2}{2k_v} + \frac{T_2 - T_1}{S} x + \frac{G_{DTV} S}{2k_v} x \quad (3A)$$

The appropriate relation for the redistribution time constant τ is Eq.(25)

$$\tau = \frac{(\ell_{10} - L/2) \rho_s h_s}{q_{\ell_1}} \quad (4A)$$

q_{ℓ_1} is the heat flux at x_1 . Specifying x_1 to be zero, taking the derivative of Eq.(3A) and evaluating $-k_s(dT/dx)$ at $x_1 = 0$ to obtain q_{ℓ_1} gives

$$q(0) = k_v \frac{(T_1 - T_2)}{S} - \frac{G_{DTV} S}{2} \quad (5A)$$

This shows that the effect of heat generation in the vapor is proportional to one half the heat release to the vapor from T_1 to T_2 . The magnitude of the error introduced will depend on the density (temperature) of the vapor and the heat flow rate $q(0)$ from the lower DT layer. For example, with an average vapor temperature of 19.5 K, $G_v S/2 = 0.0924 \text{ W/m}^2$. Although $q(0)$ will depend on the ^3He level and capsule wall, it will usually be close to the heat released from the DT displaced from the upper to the lower layer arbitrarily specified as 0.060 mm in our

discussion). This yields $q(0) = G_{DT} \times 0.060 \times 10^{-3} = 2.946 \text{ W/m}^2$. $G_v S/2$ divided by $q(0)$ then indicates an error on the order of 3%.

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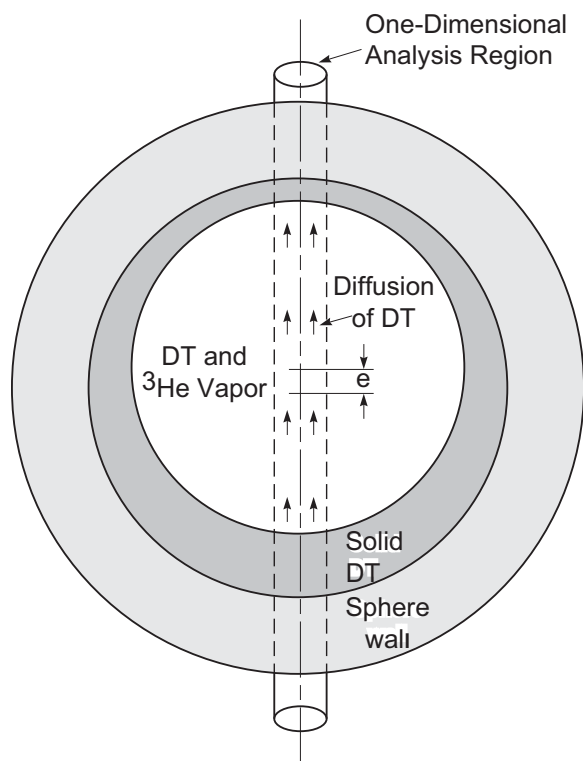


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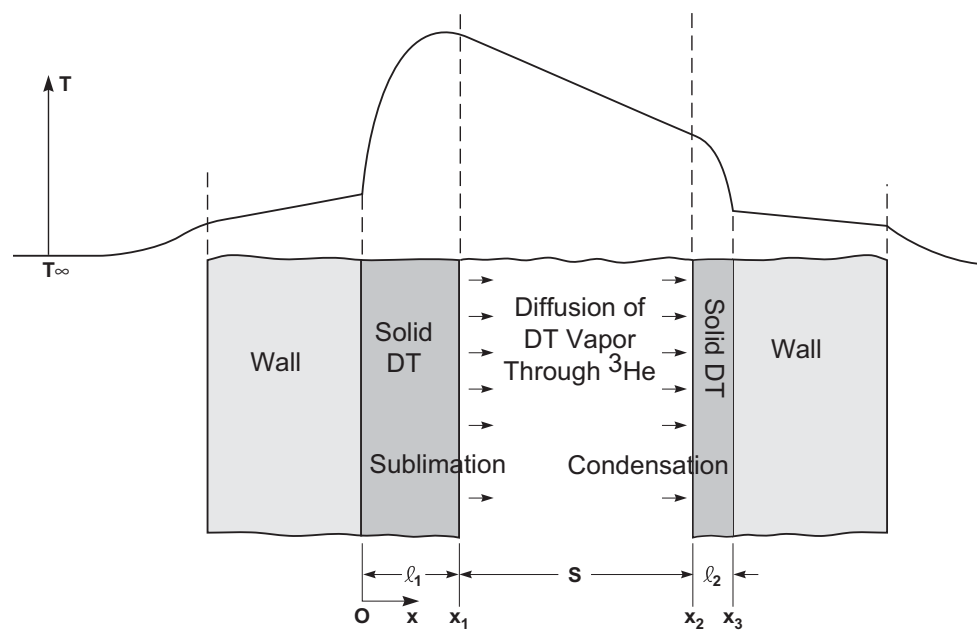


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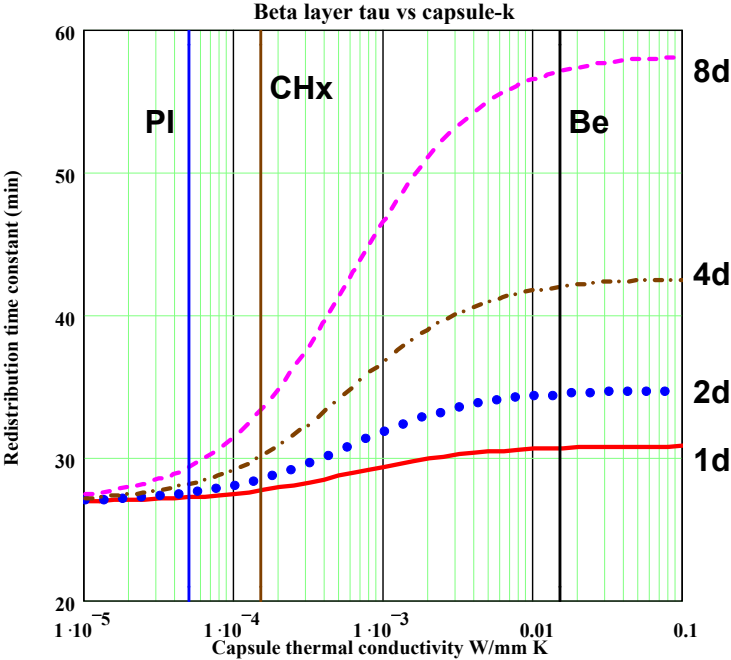


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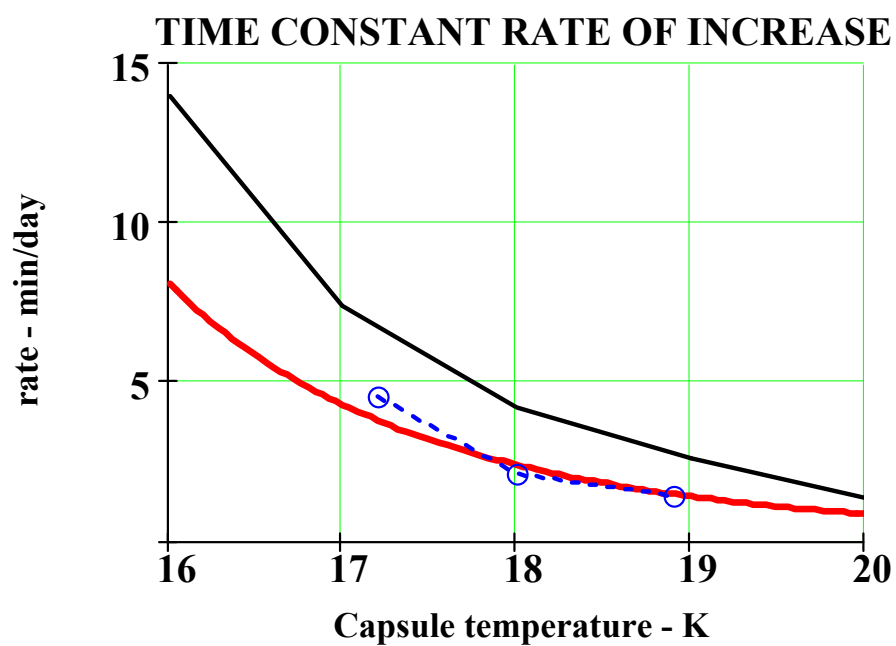


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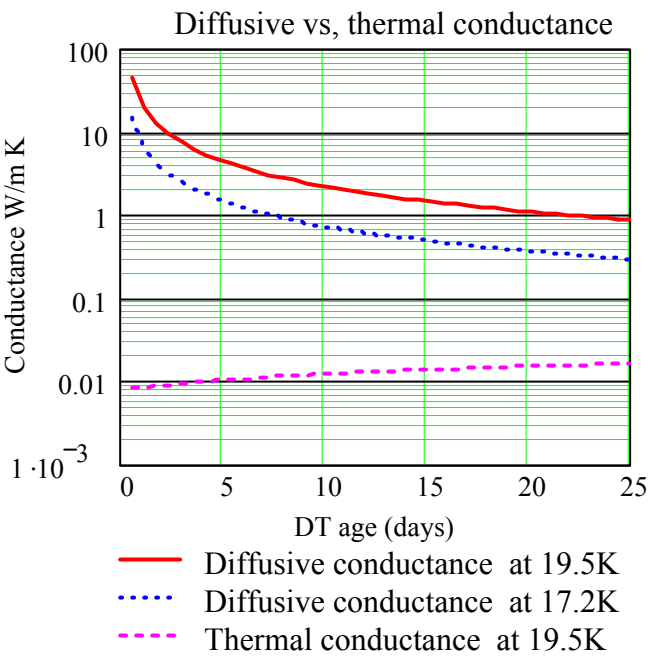


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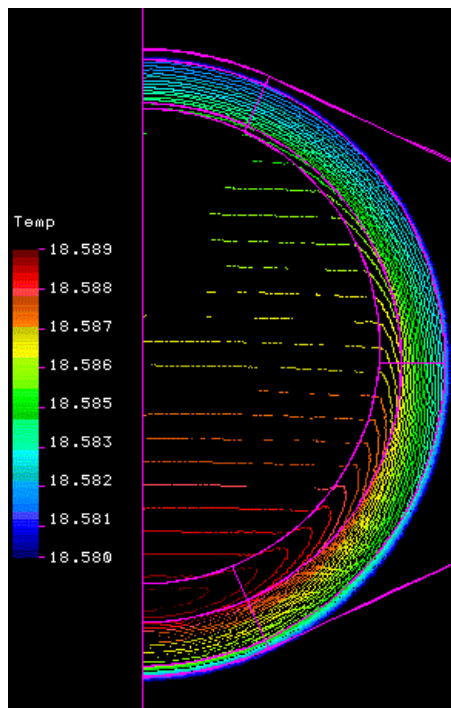


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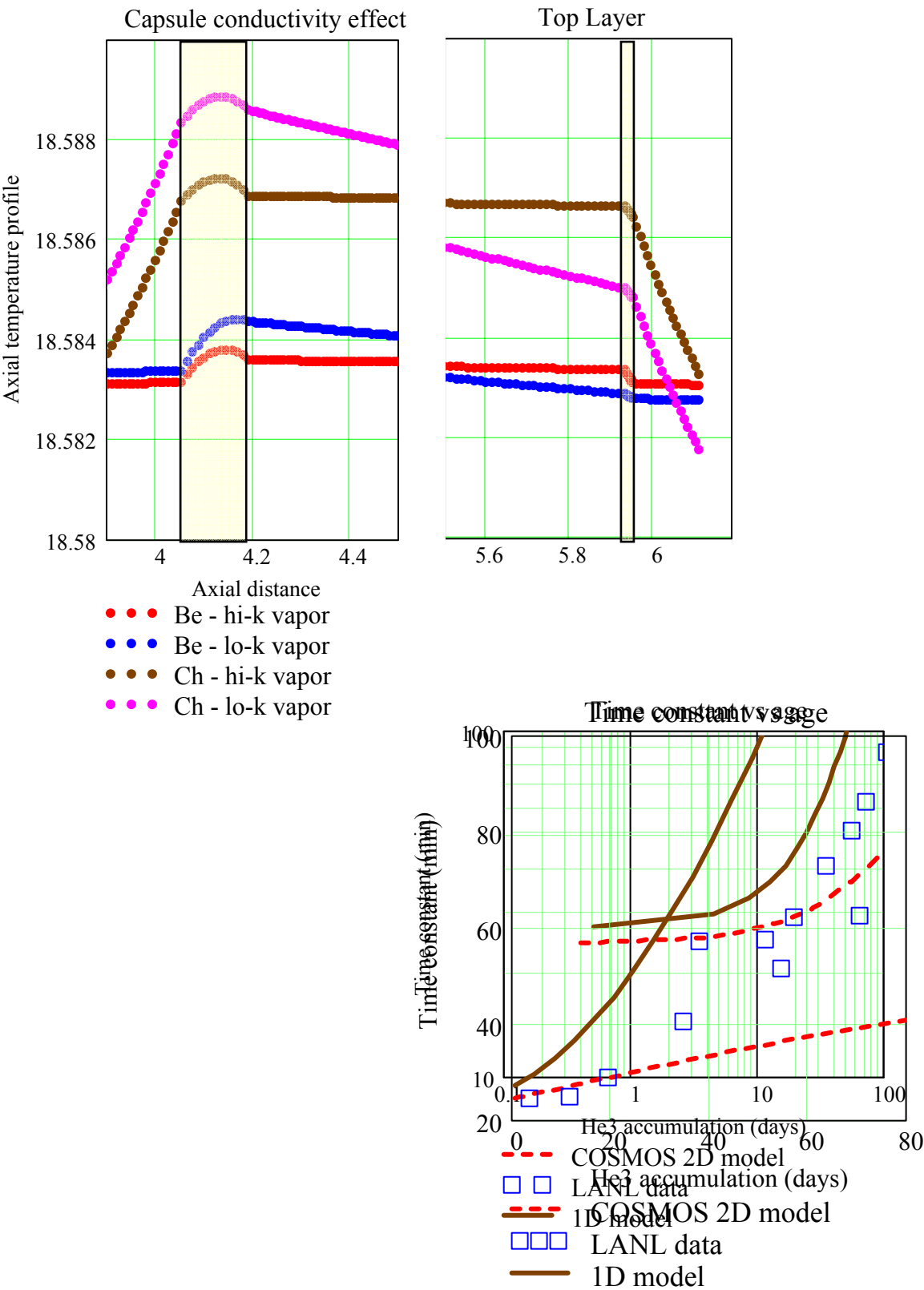


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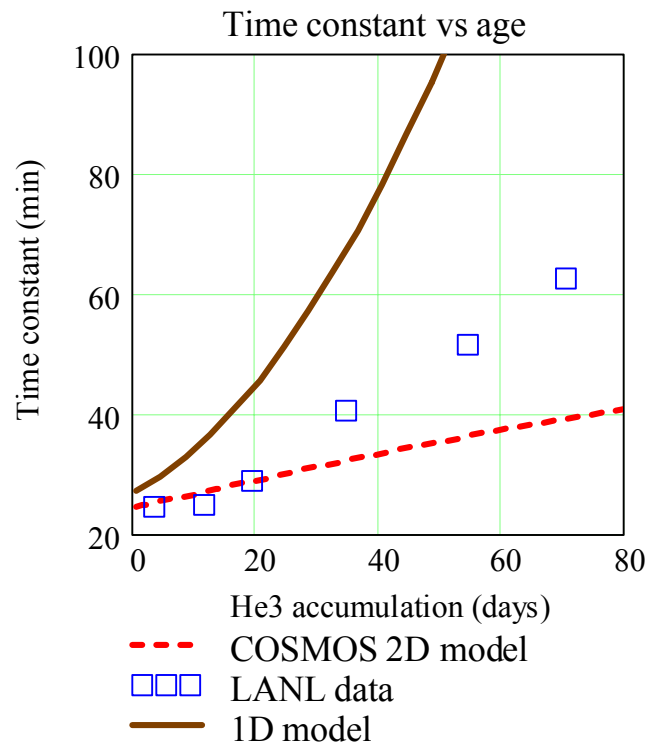


Figure A-1. Variation of diffusion coefficients with temperature for DT diffusing through ^3He .

